(NASA-CR-175880) MORPHOLOGICAL STABILITY AND FLUID DYNAMICS OF VAPOR CRYSTAL GROWTH Semiannual Progress Report, 1 Jun. - 30 Nov. 1984 (Utah Univ.) 5 p HC A02/MF A01 CSCL 20D G3/76

N85-28823

Unclas 21428

Semi-Annual Progress Report

NASA Research Grant NSG-1534

MORPHOLOGICAL STABILITY AND FLUID DYNAMICS OF VAPOR CRYSTAL GROWTH

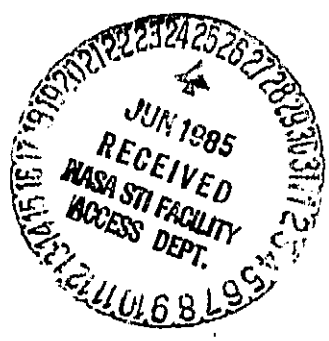
Period: 6-1-84 to 11-30-84

Franz E. Rosenberger

Department of Physics

University of Utah

Salt Lake City, UT 84112



The work performed and progress achieved during the report period, June through November 1984, are as follows:

1. Interfacial Heat and Mass Transfer

The experimental work on the thermal deflection spectroscopy measurements of the concentration field about a growing crystal was somewhat impeded by the low power output and associated instability of our argon ion laser; see the equipment item in our proposed budget for 1984 - 1985. To optimize the quantitative studies to be performed on receipt of the rebuilt laser, we have developed a rather general theoretical treatment of the thermal deflection technique. Going beyond existing treatments we have formulated the model for normal orientation of pumping and probing beams and have included the effects of the chopping frequency of the pumping beam with unequal on- and off-periods. A factor of around three can be gained in sensitivity, if a certain on-period is followed by a longer off-period. As one can intuitively expect, this leads to better cooling between heat pulses and, thus to steeper temperature gradients about the pumping beam. This, in turn, results in a steeper refractive index gradient and stronger deflection of the probing beam. These theoretical expectations were semi-quantitatively confirmed. A quantitative test will be performed as soon as the argon ion laser will be equipped with a replacement tube.

2. Interface Kinetics and Morphology

The finding of surface roughening as a precursor to a solid-solid phase transition refered to in the last progress report has been further quantified. Through improvements in the temperature stability of our microscope hot-stage and in the photographic recording process we were able to show that temperature changes as small as 0.02 ^{o}C lead to successive rounding and sharpening of corners formed by facets! A letter publication on this work has been accepted by the Journal of Crystal Growth.

In connection with the above study of the roughening of crystal corners we have discovered that such roughening can also be observed at somewhat higher temperatures, yet clearly below the phase transition, at grain boundaries. Quantification of this fundamentally and practically important observation will require more work.

Our theoretical work on the surface roughening of solid-vapor surfaces has also progressed considerably. In response to the harsh criticism of one reviewer of our original manuscrip⁺, we are in the process of including next-nearest neighbor interaction into the statistical model of surface roughening. This inclusion further increases the roughening effect of surface relaxation for copper. Application to zinc, and the most recently investigated lead, will follow shortly and hopefully unimpeachably demonstrate the validity of our model.

3. Mass Spectroscopy

Considerable progress was obtained with the mass spectroscopic characterization of the GeSe-GeI_4 vapor growth system, that was earlier flown in space. The newly designed high-temperature effusion attachment for the mass spectrometer has resulted in the desired signal enhancement. Wiedemeier et al. have postulated that the transport of GeSe is governed by the coupled reactions

$$GeSe(s) + GeI_4(v) = GeI_2(v) + 1/2\ Se_2(v) \qquad (1)$$

$$GeSe(s) = GeSe(v)\ . \qquad (2)$$

We find that the partial pressure of GeSe, when normalized with the calculated partial pressures of GeI_4 and GeI_2, agrees well with these predictions. However, the partial pressure of Se_2 lies at least two orders of magnitude lower than expected. Thus, with high probability, (1) plays no significant role in the vapor transport of GeSe. This result is further complicated by the finding that (yellow) $GeSe_2$ (also identified by powder diffraction) forms under typical transport conditions in large amounts. This compound had not been

considered in the interpretation of the space experiments. According to French and Russian phase equilibrium studies one would not expect dissociation of GeSe(s) to $GeSe_2$(s) at the temperatures used experimentally, and considered in both our work and Wiedemeier's. This appears to tie the existence of $GeSe_2$(s) to the interactive presence of iodine. Hence, future work will center on the conditions necessary for the observed $GeSe_2$ formation.

4. Vapor Transport Experiments and Calculations

Our efforts to determine a series of definitive physical vapor transport rate data that can be readily compared with theoretically obtained transport rates yielded some surprises. At total pressures above about 50 torrs, where convection is significant, the actual (3-D) rates fall below the 2-D predictions, as expected. Around 15 torrs, where diffusion dominates, good agreement between experiments and model predictions is obtained. However, at lower pressures the actual rates exceed the predictions considerably. Any attempts to interpret this unexpected deviation in terms of a systematic experimental error have failed so far. We will now conduct a series of experiments at various low pressures to explore the pressure dependence in this range.

5. Recent Publications and Presentations of NASA-Sponsored Research

Publications

N.-B. Ming, J.-S. Chen and F. Rosenberger: Surface Roughening as a Precursor to the Polymorphic Phase Transition in CBr_4. J. Crystal Growth 69, 631 (1984).

C. Smutek, P. Bontoux, B. Roux, G. H. Schiroky, A. C. Hurford, F. Rosenberger and G. de Vahl Davis: Three-Dimensional Convection in Horizontal Cylinders: Numerical Solutions and Comparison with Experimental and Analytical Results. Numerical Heat Transfer (submitted).

P. Bontoux, B. Roux, G. H. Schiroky, B. Markham and F. Rosenberger: Analytical

and Two-Dimensional Approximations for Convection in the Vertical Midplane of a Horizontal Cylinder. Int J. Heat Mass Transfer (submitted).

Oral Presentation

The Absence of Convective Stability in Incongruent Crystal Growth on Earth. Twenty-fifth COSPAR Meeting, Graz, Austria, July 2-5, 1984.

Recognition

Chairman, Gordon Research Conference on Crystal Growth, Plymouth, N. H., July 15-19, 1985.